August 3, 2007

Randy Segawa
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Dear Mr. Segawa:

I appreciate the opportunity to provide a peer review of the Proposed Regulations for the Reduction of the Emission of Volatile Organic Compounds. A copy of my review comments is enclosed. Please contact me if any of these points requires clarification, if I have neglected to address an important issue, or if you have any other concerns.

I will destroy all documentation provided with the review request, as per an e-mail from Pam Wofford dated August 2, 2007.

Sincerely,

ORIGINAL SIGNED BY

SHARON K. PAPIERNIK
Research Soil Scientist

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Peer review comments
Proposed Regulations for the Reduction of the Emission of Volatile Organic Compounds

A. General comments
1. It is counterintuitive that fumigants, which are highly volatile, are the only pesticides for which VOC production is assumed to be less than 100%. In three of the five nonattainment areas, estimated VOC emissions from "other pesticides" exceed 2004 fumigant emissions. Cumulative volatilization loss of non-fumigant pesticides is rarely more than 50% of the parent compound. For soil- and foliar-applied pesticides of relatively low volatility, volatilization is typically less than 20%. Even though pesticide degradation products may also contribute to VOCs, it is unlikely that an emission rate of 100% is reasonable for non-fumigant pesticides. Glotfelty et al. (1989) presented an empirical equation that provides a first estimate of volatilization rates based on the physicochemical properties of the pesticide; other predictive equations have been published for soil- and foliar-applied pesticides (for example, Haith et al., 2002; Voutas et al., 2005).

2. It is not clear why field studies using flux chambers were excluded from the database. While flux chambers can produce inaccurate absolute values for fumigant volatilization, comparisons between management factors may be relatively robust. Aeordynamic methods are also subject to error, depending on the sampling period and other factors (Majewski, 1996). A recent analysis by van Wesenbeeck et al. (2007) indicated good agreement in fumigant flux measured using dynamic flux chambers and aerodynamic methods. In these proposed regulations, only one adjustment factor is used for each management option, with no regard for application depth, temperature, antecedent soil moisture, etc. It would seem reasonable that flux chamber data could provide valuable information for the development of these adjustment factors without compromising the integrity of the values.

3. Soil column data (Gan et al., 1997) are accepted to estimate the effect of application methods on volatilization of methyl bromide, but similar soil column experiments are not mentioned for 1,3-D, chloropicrin, or MITC volatilization. Many such column experiments have been conducted, as discussed in more detail in the fumigant-specific sections below. While these column studies may not accurately predict fumigant emissions under field conditions, they may provide valuable information for comparing the effects of management practices on potential emissions. Experimental results from laboratory soil columns should either be used uniformly as supporting evidence or excluded for all fumigants.

4. Better definitions of tarp permeability are needed. In these proposed regulations, tarps have an upper and lower limit on their permeability, as stated for methyl bromide in section 6447(e). Similar definitions of tarp permeability are not provided for the other fumigants. Papiernik et al. (2001) present a method for describing the permeability of agricultural films that does not depend on the concentration gradient across the film. This mass transfer approach could eliminate some ambiguity in the designation of film permeability. The mass transfer coefficient is a property of the film-chemical combination that increases with increasing temperature, but is otherwise unaffected by environmental conditions (Papiernik and Yates, 2002).

5. Field studies have consistently shown that use of a virtually impermeable film can reduce emissions of all fumigants to less than one-third that from HDPE-tarped soil, provided the film remains intact for a sufficient length of time (Wang et al., 1997; Papiernik et al., 2004b; Gao and Trout, 2007; van Wesenbeeck et al., 2007). These films are especially effective if a continuous soil cover can be achieved, as in broadcast fumigation. It appears that these proposed regulations elect not to include VIF because some researchers and growers have experienced problems with film tearing and ineffective seam gluing. These problems may have been addressed by the film manufacturers. Potential restrictions on soil fumigation in some non-attainment areas may justify the significant added expense of a VIF to reduce fumigant emissions. Inclusion of VIF as an approved tarp should be considered, using cover times of 10 days or more.

6. The amount of water recommended for water sealing is quite low compared to the amount typically added in research studies. The proposed regulations specify three applications of 0.25 inches of water. The water sealing approaches outlined in field research studies typically use 0.5 inches or more water for at least one of the water applications for metam sodium (Sullivan et al., 2004), and 1,3-dichloropropene plus chloropicrin (Gao and Trout, 2007) fumigation. The registrant's study included in the dazomet documentation used irrigations of greater than 0.25 inches in the first 2 days after application. Thus, it is not known whether the proposed adjustment factors will be appropriate for the low water amounts required in the proposed regulations.

7. Adjustment factors were usually determined by grouping field studies by application method and averaging the cumulative emissions. This is a simple, straightforward way of calculating an average emission for a management practice. In many cases, the variation in measured emissions is very large (CV >30%). The validity of using an average value depends on the intentions of DPR in making these determinations. Assuming that all studies are equally accurate in their volatilization measurement and in their representation of commercial fumigation practices (I have little information to support or refute that assumption), using the average will tend to equally underpredict and overpredict actual emissions. If the proposed regulations are intended to reflect conditions under which emissions might be greater than the average, then a higher value should be used. It would be more conservative to use the maximum measured emissions as an adjustment factor, but this will likely overestimate emissions using current fumigation practices.

In the case of 1,3-dichloropropene, emissions were determined from four field studies in which 1,3-D was applied at four different depths. Emissions were interpolated to estimate emissions from two uniform application depths (12 and 18 inches). It would be more straightforward to use the two shallow applications to derive the 12-inch application value (65%) and the two deep applications to derive the 18-inch application value (26%). As noted in the documentation, the deep application studies were conducted under unrepresentatively cool conditions, and higher emissions are expected during a typical California fumigation. For the purposes of this analysis, the linear interpolation used to determine adjustment factors for 1,3-D shank application are reasonable and the resulting factors are consistent with field observations.

The field studies used to determine adjustment factors were conducted under a variety of conditions that may not always be representative of typical California soil fumigations. I recognize that insufficient evidence exists for the development of seasonal adjustment factors,

etc. It is understood that these adjustment factors will be changed as more information becomes available regarding VOC emissions from pesticide applications. The GLP studies approved by the DPR are the best source of information for the development of adjustment factors, but these studies do not provide a complete data set. Below, I outline additional information supporting some proposed adjustment factors, and point out studies that provide information indicating that some proposed adjustment factors may need to be reevaluated.

B. Methyl Bromide
The adjustment factor of 74% for bare soil appears reasonable, based on available data, but it should be recognized that methyl bromide emissions measured in field studies have been highly variable, and cumulative emissions of nearly 100% have been reported. Volatilization measured in untarped laboratory soil columns totaled 37 to 82% of the applied methyl bromide under a wide variety of soil conditions (Gan et al., 1996, 1997, 1998a, 1998b), supporting the proposed adjustment factor for 1990-1991 applications.

The conclusion that broadcast fumigation with MeBr using a HDPE (low permeability) tarp will produce emissions of 48% is not well-supported by field data. The assumption that total emissions are twice the 24-hour emissions requires further substantiation when field studies (such as Yates et al., 1997) and lab studies (Gan et al., 1997) have shown that this may not be true in all situations. A review of many field experiments monitoring methyl bromide volatilization indicates that emissions from HDPE-tarped soil are usually >50% of the applied methyl bromide (Yates et al., 2003). An analysis of the results of field studies published in the peer-reviewed literature indicates that the overall mean emissions from HDPE-tarped soil total 52% of the applied methyl bromide, but measured emissions range from <30% to >80% (Yates et al., 1998). The proposed adjustment factor, in my opinion, is not conservative enough to account for the large fraction of fumigations completed under conditions resulting in methyl bromide emissions significantly greater than 48%. For some of the field studies summarized in Table 1, peak emissions of >40% were measured from HDPE-tarped soil within the first 24 hours after methyl bromide application, indicating that total emissions were likely much greater than 48%.

The assumption that emissions will be high in bedded systems is supported by the observations of Wang and Yates (1998), who determined high methyl bromide loss (~95%) from beds partially covered with plastic film (leaving the furrows bare).

The assumption that deep applications have the same emissions as shallow applications is reasonable, but limited research has shown that increasing the depth of application can reduce cumulative emissions in some cases (Yates et al., 1997; Gan et al., 1997).

The assumption that structural fumigation will result in 100% emissions is reasonable, if the controlled atmosphere is released with no treatment.

C. 1,3-Dichloropropene
Adjustment factors for shank application are based on four field studies. These values, which relied on interpolation of the results from each of these four studies to two uniform application depths (12 and 18 inches), appear reasonable and are supported by other studies published in the

peer-reviewed literature. The adjustment factors for shallow injection (61%) and deep injection (41%) are consistent with field and laboratory studies. Cryer et al. (2003) report cumulative volatilization rates of ~40% for shank bed injection at a depth of 10 inches for a field study conducted in Florida. Laboratory soil column experiments (Zheng et al., 2006b) using bare soil indicated that cumulative 1,3-D emissions were 65% for shank injection at a depth of ~8 inches and 46% for shank injection at ~16 inches. Soil column experiments for 1,3-D injection at a depth of 12 inches (no tarp) produced cumulative volatilization of ~40% (Gan et al., 2000a); injection at 18 inches produced cumulative volatilization of 33% in an unamended soil (Ashworth and Yates, 2007).

No adjustment factor is provided for "low-permeability" tarps. Field studies using flux chambers measured a 27% reduction in cumulative emissions (relative to bare soil) when the soil was covered with a polyethylene tarp (Gan et al., 2000a). Studies using laboratory soil columns indicate that HDPE is relatively ineffective in reducing 1,3-D emissions, with cumulative emissions >85% those from similar untarped columns (Gao and Trout, 2006; Gan et al., 1998c). Use of the same adjustment factor for high- and low-permeability tarps (or no tarp) may be reasonable.

No basis is given for the assumptions regarding surface water application (cumulative emissions approximately 67% of the untarped soil value). Gao and Trout (2007) used flux chambers, and had some problems with sealing the chamber, as noted in the DPR documentation. They applied water six times after application, and observed that 24% of the applied 1,3-D volatilized during the experiment. Average emissions from water-sealed soil were 73% of that measured from HDPE-tarped soil. Using soil columns and surface water application similar to that required in the proposed regulations, Gao and Trout (2006) found that such water sealing was not particularly effective in reducing emissions. Cumulative emissions from soil columns receiving three water treatments were similar to cumulative flux from HDPE-tarped soil (41% versus 44% of applied) when 1,3-D was injected at a depth of 12 inches (Gao and Trout, 2006). Another experiment using soil columns (Ashworth and Yates, 2007) found that by irrigating daily for the first five days following fumigant application, cumulative emissions were reduced by about 50% relative to bare, nonirrigated soil. The timing of water application and the amount of water added are expected to have a very large influence on the effectiveness of surface water application in reducing 1,3-D emissions. As noted in the "General comments" section above, these experiments generally applied more water more frequently than the water application required in the proposed regulations, so experimental results may underestimate the expected emissions under conditions following the proposed regulations.

Use of a single adjustment factor for drip application may need to be reevaluated. Emissions of 1,3-D measured by van Wesenbeeck et al. (2007) following drip application to tarped, bedded soil in Georgia (average 24% emissions) and Florida (average 21% emissions) were consistent with the proposed adjustment factor of 29% for drip application. Field studies using flux chambers indicated cumulative emissions of ~32% for subsurface drip application at a depth of 4 inches (Gan et al., 2000a), also consistent with the proposed adjustment factor. In that study, cumulative 1,3-D emissions following subsurface drip application were about 70% of those resulting from shank injection to HDPE-tarped soil beds. Similar cumulative emissions for subsurface drip (22% loss of 1,3-D) were observed in soil column experiments (Gan et al.,

1998c). Wang et al. (2000) measured substantially higher 1,3-D emissions (57%) when the fumigant was applied by subsurface drip at an 8-inch depth. Surface drip application of fumigants is expected to result in very high volatilization rates under some conditions. While van Wesenbeeck et al. (2007) measured relatively low emissions from surface drip application of 1,3-D to tarped beds in their Georgia study (24%), Wang et al. (2000) measured high emissions (66%). Laboratory soil column studies measured >90% cumulative emissions in surface drip 1,3-D applications to bare soil (Gan et al., 1998c). Papiernik et al. (2004a) measured low 1,3-D emissions from HDPE-tarped beds with subsurface drip-applied 1,3-D under cool conditions; they noted that increasing the depth of subsurface drip application decreased cumulative emissions. The proposed regulations (6448.1) do not include a drip application with no tarp. The proposed adjustment factors should specify subsurface drip application and/or use of a HDPE tarp for drip application.

D. Chloropicrin
As pointed out in the DPR documentation, information is sparse regarding the volatilization of chloropicrin after soil application. In addition to the studies submitted to DPR on behalf of the registrant, only a few studies appear in the peer-reviewed literature. Gao and Trout (2007) experienced problems sealing their flux chambers to the bare soil, and they measured low chloropicrin emissions (<10% of applied) for all application methods. Gan et al. (2000b) used laboratory soil columns to evaluate the impact of surface tarps on chloropicrin volatilization. Cumulative chloropicrin emissions were from bare soil 82%, and emissions from HDPE-tarped soil were approximately one-fourth that (20% of applied). Chloropicrin generally degrades in soil quite rapidly, and soil conditions can have a large impact on its environmental fate (Gan et al., 2000b). Thus, volatilization of chloropicrin is expected to be highly variable, depending on the conditions prevailing during the fumigant application. In addition, chloropicrin degrades rapidly in the presence of metam sodium (Zheng et al., 2004). Flux studies which include mixtures of chloropicrin and metam sodium (for example, Wang et al., 2005) may not produce accurate predictions of chloropicrin emissions when chloropicrin is applied alone.

No experimental evidence is cited to support the proposed adjustment factor for surface water application. Wang et al. (2005) observed higher cumulative emissions of chloropicrin in water-sealed soil (irrigated daily for 7 days after application) than in HDPE-tarped soil, contrary to the proposed adjustment factors. As noted above for 1,3-D, the effectiveness of this management practice is expected to be highly variable, depending on the intensity and timing of water application.

Only one study using drip application of chloropicrin is cited in the documentation. van Wesenbeeck et al. (2007) measured emissions of chloropicrin following drip application to tarped, bedded soil in Georgia (average 18% emissions) and Florida (average 32% emissions), both higher than the proposed drip adjustment factor of 15%.

E. Metam sodium and metam-potassium
Limited information is available regarding emissions of MITC from fields fumigated with metam products. Emissions of MITC are highly variable, and the proposed adjustment factors for applications of metam with no surface treatment (77% emissions) appear reasonable. Field studies have measured cumulative MITC emissions of ~1% to >80% of the applied metam

sodium (Sullivan et al., 2004; studies cited in DPR documentation). Laboratory soil column experiments simulating shank injection to untarped soil have measured MITC cumulative emissions amounting to a few percent (Frick et al., 1998) to >50% of the applied chemical (Gan et al., 1998b; Zheng et al., 2006a). Some of these laboratory studies injected MITC directly, and so do not account for the conversion of metam to MITC.

Some field monitoring has suggested that air concentrations of MITC (and cumulative emissions) might be lower for shank injection than for rotovator (van den Berg et al., 1999) or sprinkler application (Saeed et al., 2000). The low adjustment factors for rotovator and soil capping may require additional justification.

Field studies using flux chambers have measured low cumulative emissions of MITC (<2%) following subsurface drip application to HDPE-tarped beds under cool conditions (Papiernik et al., 2004a), in agreement with the proposed DPR adjustment factors. Li et al. (2006) also report cumulative emissions of <3% for metam sodium applied via surface drip to tarped beds. Laboratory soil columns experiments have shown large potential emissions from surface-applied metam sodium to bare soil: Zheng et al. (2006a) measured cumulative emissions of 80%, in agreement with the proposed adjustment factors for sprinkler and flood application. Similar emissions are expected for surface drip application with no tarp, and unless DPR has supporting data, the proposed adjustment factors should specify subsurface drip application and/or a HDPE tarp for these low adjustment factors (9%).

As noted above, research results evaluating the impact of water sealing on MITC emissions are not directly comparable to the proposed regulations, because research studies typically apply much more water than is required in the proposed regulations. Sullivan et al. (2004) note the requirement for nighttime water application for reducing MITC emissions. The water application requirements outlined in the proposed regulations (6450.1) may not be sufficient to achieve the emissions reductions specified by the proposed adjustment factors.

F. Dazomet
The studies cited in Table 5 indicate that emissions of MITC following dazomet application are highly variable. Wang et al. (2005) used passive flux chambers to measure MITC volatilization resulting from dazomet application. Cumulative MITC emissions totaled <5% of the applied chemical (assuming 100% conversion of dazomet to MITC) for both HDPE-tarped and water-sealed soil, supporting the low adjustment factor (17%) proposed for dazomet applications.

G. Sodium tetrathiocarbonate

I am not aware of any detailed emissions studies for carbon disulfide other than the study provided with the proposed regulation documentation. The proposed adjustment factors (10%) do not appear to be conservative, based on the observed emissions (9.6%) and the limited length of time during which CS_2 concentrations were monitored, in which CS_2 flux was still increasing when monitoring was terminated.

H. Other pesticides
Discussed above in "General comments" section.

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